

RECEIVED
CENTRAL FAX CENTER
DEC 04 2007

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: :
H. Nabeta et al. : Art unit: 2814
Serial No.: 10/590,589 :
Filed: August 24, 2006 :
For: WHITE LIGHT EMITTING :
DIODE (WHITE LED) AND :
METHOD OF MANUFACTURING: :
WHITE LED :

DECLARATION

Honorable Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

I, Hideaki KARAMON hereby declare and say as follows:

I am familiar with both the English and Japanese languages and I have compared the annexed English translation with the Japanese text of Japanese Patent Application No. 61931/2004.

To the best of my knowledge and belief, the annexed English translation is an accurate translation of the above Japanese application.

The undersigned declares further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the

knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both under Section 1001 of Title 18 of the U.S. Code and that such willful false statements may jeopardize the validity of the above-identified application or any patent issuing thereon.

A handwritten signature in black ink, appearing to read 'H. Karamon', is written over a horizontal line.

Hideaki KARAMON

Dated: This 4th day of December, 2007.

RECEIVED
CENTRAL FAX CENTER
DEC 04 2007

Patent Application No. 61931/2004

Title of the Document: APPLICATION FOR PATENT

Reference Number: OH0018328

Address: The Director General of the
Patent Office

International
Patent Classification: H01L 33/00
C09K 11/08

Inventor(s):

Post-Office Address: C/O Konica Minolta Technology
Center, Inc.
1 Sakura-machi, Hino-shi,
Tokyo, Japan

Name: Hiroyuki NABETA

Post-Office Address: C/O Konica Minolta Technology
Center, Inc.
1 Sakura-machi, Hino-shi,
Tokyo, Japan

Name: Hideaki WAKAMATSU

Applicant for patent:

Identification Number: 000001270

Name: Konica Minolta Holdings, Inc.

Representative: Fumio IWAI

Indication of Fee:

Prepayment Registration
Number: 012265

Amount of Payment 21000

List of Documents Attached:

Title of Document:	Scope of Patent Claims	1
Title of Document:	Specification	1
Title of Document:	Drawings	1
Title of Document:	Abstract	1

[NAME OF THE DOCUMENT] SCOPE OF PATENT CLAIMS

[Claim 1] A white light emitting diode comprising a phosphor layer to convert blue light into yellow light, provided on a blue light emitting diode,

wherein the phosphor layer comprises a phosphor.

[Claim 2] A white light emitting diode comprising a phosphor layer to convert blue light into yellow light, provided on a blue light emitting diode,

wherein the phosphor layer comprises a phosphor and a transparent inorganic oxide.

[Claim 3] A method of manufacturing the white light emitting diode of Claim 1, comprising the step of:

colliding phosphor particles with the blue light emitting diode at high speed to form the phosphor layer on the blue light emitting diode.

[Claim 4] A method of manufacturing the white light emitting diode of Claim 2, comprising the step of:

colliding phosphor particles and transparent inorganic oxide particles with the blue light emitting diode at high speed to form the phosphor layer on the blue light emitting diode.

[NAME OF THE DOCUMENT] SPECIFICATION

[TITLE OF THE INVENTION] WHITE LIGHT EMITTING DIODE (WHITE LED) AND METHOD OF MANUFACTURING WHITE LED

[FIELD OF THE INVENTION]

[0001]

The present invention relates to a white light emitting diode (white LED) in which blue light emitted from a blue source; particularly a blue LED element is converted into white light, and a method of manufacturing a white LED.

[BACKGROUND OF THE INVENTION]

[0002]

In recent years, a white LED has been brought to attention as a white lighting light source exhibiting high efficiency as well as high reliability, and has already been utilized partly as a small-size low electric power light source. It is commonly known that this kind of LED is a LED in which a blue LED element is covered by a mixture of a yellow phosphor and a transparent resin, and a white LED and a phosphor used for the white LED with this method are disclosed (refer to Patent Documents 1 - 3).

[0003]

However, a blue light tends to deteriorate a resin since energy of the blue light is large. Therefore, the

resin loses the color by using the white LED having the above-described structure for long duration, whereby the color tone is also changed. Further, in recent years, though there is a move to introduce the development of a white lighting light source by employing a high-energy LED element, the resin is seriously deteriorated in this case since a limited area is exposed to extremely large energy blue light, whereby change of an emission color is caused in an extremely short period of time. There was also a problem such that the emission color tone was shifted to the yellow side, since a heat discharge property of a resin-molded element is poor, whereby temperature rise is easily generated.

(Patent Document 1) Japanese Patent O.P.I. Publication
No. 10-163535

(Patent Document 2) WO98/05078; descriptive literature

(Patent Document 3) Japanese Patent O.P.I. Publication
No. 2002-43624

[DISCLOSURE OF THE INVENTION]

[PROBLEMS TO BE SOLVED BY THE INVENTION]

[0004]

The present invention was made on the basis of the above-described situation.

[0005]

It is an object of the present invention to provide a white LED exhibiting high reliability and longer operating life, which is prepared via use of a blue LED element; particularly a high-energy blue LED element, and a manufacturing method of the same.

[MEANS TO SOLVE THE PROBLEMS]

[0006]

After considerable effort during intensive studies, the inventors have found out that the foregoing object can be accomplished by the following structures.

[0007]

(Structure 1) A white light emitting diode comprising a phosphor layer to convert blue light into yellow light, provided on a blue light emitting diode, wherein the phosphor layer comprises a phosphor.

[0008]

(Structure 2) A white light emitting diode comprising a phosphor layer to convert blue light into yellow light, provided on a blue light emitting diode, wherein the phosphor layer comprises a phosphor and a transparent inorganic oxide.

[0009]

(Structure 3) A method of manufacturing the white light

emitting diode of structure 1, comprising the step of colliding phosphor particles with the blue light emitting diode at high speed to form the phosphor layer on the blue light emitting diode.

[0010]

(Structure 4) A method of manufacturing the white light emitting diode of Structure 2, comprising the step of colliding phosphor particles and transparent inorganic oxide particles with the blue light emitting diode at high speed to form the phosphor layer on the blue light emitting diode.

[EFFECT OF THE INVENTION]

[0011]

A white LED exhibiting high reliability and longer operating life can be provided by utilizing a blue LED.

[DETAILED DESCRIPTION OF THE INVENTION]

[0012]

Next, a white light emitting diode (white LED) of the present invention, and a method of manufacturing a white LED in the present invention will be further described in detail.

[0013]

Fig. 2 is a schematic cross-sectional view showing white LED 21 of the present invention. After forming bumps 25 on the surface of blue LED chip 24, blue LED chip 24 is

turned over to be connected to electrodes 23 on substrate 22 (so-called flip-chip bonding). Phosphor layer 26 is further formed on the blue LED by a film formation method in which phosphor particles of the present invention are deposited via high-speed collision of the particles with the chip surface. Sealing layer 27 made of transparent inorganic oxide may further be formed on phosphor layer 26 as shown in Fig. 2.

[0014]

Any of known blue LEDs including an $\text{In}_x\text{Ga}_{1-x}\text{N}$ type LED is usable as a blue LED employed for the present invention. It is preferred that a blue LED has a light emission peak wavelength is 440 - 480 nm.

[0015]

A commercially available phosphor capable of converting blue light emitted from a blue LED into yellow origin light which is greenish yellow (a light emission wave length of about 550 nm), for example is usable as a phosphor employed for the present invention. $\text{Y}_3\text{Al}_5\text{O}_{12}$ based phosphor such as (Y, Gd, Ce) $_3\text{Al}_5\text{O}_{12}$ and the like is employed as the most preferable oxide phosphor.

[0016]

A transparent inorganic oxide other than the foregoing phosphor may be mixed in a phosphor layer of the present

invention. SiO_2 , Al_2O_3 and so forth are provided as a transparent inorganic oxide usable for the present invention.

[0017]

(Process of forming a phosphor layer)

Utilized is a film formation method (so-called aerosol deposition method) in which particles of phosphor or transparent inorganic oxide as a raw material are deposited on a blue LED as a substrate via high-speed collision of the particles with the chip surface, in order to form a phosphor layer in the present invention.

[0018]

An aerosol deposition film-forming apparatus disclosed in "OYO-BUTURI" (Vol. 68, No. 1, Page 44) and Japanese Patent O.P.I. Publication 2003-215256 can be utilized.

[0019]

Fig. 1 shows a schematic diagram of an aerosol deposition film-forming apparatus used in the present invention. The aerosol deposition film-forming apparatus is equipped with holder 9 supporting substrate 10, XYZθ stage 11 to set the holder 9 in 3 dimensional motion, nozzle 8 having a narrow opening to spray a raw material onto the substrate, nozzle 8 introduced into chamber 7 connected to aerosol generation container 4 via pipe 6, high-pressure gas cylinder

1 to accumulate a carrier gas, aerosol generation container 4 in which particle raw material 12 and the carrier gas transferred via pipe 2 are stirred to be mixed. A temperature control system with a peltiert element is placed on the back surface of the stage to keep the substrate in optimum temperature.

[0020]

Further, the particle raw material in the aerosol generation container is formed on the blue LED as a substrate by the following procedures.

[0021]

The raw material particle filled in the aerosol generation container has a preferable particle diameter of $0.02 - 5 \mu\text{m}$, or a more preferable particle diameter of $0.1 - 2 \mu\text{m}$. The particle raw material passes through the pipe to the aerosol generation container from the high-pressure gas cylinder with the accumulated carrier gas, and the particle raw material together with the carrier gas is aerosolized via vibration while stirring.

[0022]

In order to measure a particle diameter of the raw material particle, a conventional laser diffraction type

particle size analyzer is employed. Specific examples of the laser diffraction type particle size analyzer include HELOS (manufactured by JEOL Ltd), Microtrac HRA (manufactured by Nikkiso Co., Ltd.), SALD-1100 (manufactured by Shimadzu Corp.) and Coulter Counter (manufactured by Coulter Corp.). Of these, Microtrac HRA is preferably employed.

[0023]

The aerosolized particle raw material passes through the pipe, and the particle raw material together with the carrier gas is sprayed onto the substrate from the nozzle having a narrow opening in the chamber to form a coating layer. The chamber is evacuated by a vacuum pump or such, and a vacuum degree inside the chamber is arranged to be adjusted if desired. In the present invention, the vacuum degree is preferably 0.01 - 10000 Pa, and more preferably 0.1 - 1000 Pa. Further, a phosphor layer having a desired thickness is possible to be formed at the predetermined portion of the substrate, since the substrate holder can be moved by setting the XYZθ stage in 3 dimensional motion. A sealing layer can be formed on the prepared phosphor layer, if desired.

[0024]

The aerosolized raw material particles are transported

by a carrier gas having a flow speed of 100 - 400 m/sec, and deposited via collision with the substrate surface. The particles transported by the carrier gas are also bonded via impact caused by collision with each other to form a layer.

[0025]

In the manufacturing method of the present invention, inert gas such as N_2 gas or He gas is preferable as a carrier gas to accelerate and spray the raw material particles, but N_2 gas is more preferably usable.

[0026]

It is preferred that a temperature of at least - 100 °C and at most 200 °C is also maintained as a temperature of the substrate to collide the raw material particles with. When the substrate temperature is increased to at least 300 °C, the film becomes milky-white, and light can not be taken out, whereby luminance of a white LED tends to be lowered.

[0027]

Particles of at least the foregoing phosphor are necessary to form a phosphor layer, but the foregoing transparent inorganic oxide particles may also be mixed, if desired. The aerosol generation containers for the phosphor and additionally for the transparent inorganic oxide are installed in the foregoing film-forming apparatus, and a

phosphor distribution in the phosphor layer may be controlled by changing one feed raw material to another appropriately. The phosphor concentration in the phosphor layer can be controlled by appropriately mixing a phosphor with a transparent inorganic oxide. In the case of forming only the transparent inorganic oxide layer on the outermost surface, it is usable as a transparent sealing layer. On the other hand, only the transparent inorganic oxide layer may be formed on the blue LED surface. It is also possible to form a layer composed of no transparent inorganic oxide, but only a phosphor.

[0028]

A LED chip formed with a phosphor layer is fitted with a transparent resin cover made of a silicone resin or such, or a glass cover at the phosphor film-formed portion of the light emitting chip to complete a white LED. The rated direct current load up to a maximum voltage and current of 5 V and 30 mA, respectively, for example, is applied to a white LED of the present invention to emit light, whereby white light emission can be obtained.

[EXAMPLE]

[0029]

Next, the present invention will now be described in

detail referring to examples, however, the present invention is not limited thereto.

[0030]

(Example 1)

A yellow phosphor layer was prepared employing an aerosol deposition film-forming apparatus as shown in Fig. 1. (Y, Gd, Ce)₃Al₅O₁₂ phosphor particles having a particle size distribution of 0.1 - 1 μm and an average particle diameter of 0.5 μm were filled in an aerosol generation container, and sprayed onto a blue LED chip (0.4 mm-square) having a light emission peak wavelength of 460 nm under the conditions of a chamber vacuum degree of 100 Pa and a substrate temperature of 20 °C, employing N₂ gas with a flow rate of 200 m/s as a carrier gas to prepare a 10 μm thick film, whereby a white LED was obtained.

[0031]

(Example 2)

Similarly to Example 1, (Y, Gd, Ce)₃Al₅O₁₂ phosphor particles were sprayed onto a blue LED chip (0.4 mm-square) having a light emission peak wavelength of 460 nm to prepare a 10 μm thick film. Further, SiO₂ particles having a particle size distribution of 0.1 - 1 μm and an average

12

particle diameter of 0.5 μm were sprayed onto the above-described resulting sprayed surface under the same condition to prepare another 10 μm thick film, whereby a white LED shown in Fig. 2 was obtained.

[0032]

(Comparative example)

A mixture liquid in which $(\text{Y, Gd, Ce})_3\text{Al}_5\text{O}_{12}$ phosphor was mixed with an epoxy resin (NT8014, produced by Nitto Denko Corp.) and an acid anhydride based hardener was prepared.

[0033]

Fifty μl of the above mixture liquid of the phosphor and the resin was dripped onto a blue LED chip (0.4 mm-square) having a light emission peak wavelength of 460 nm, employing an injector, and after the resulting surface was dried, it was further covered by a half-circle-shaped transparent epoxy resin cover to obtain a white LED.

[0034]

(Evaluation)

Each of white LEDs of Example 1, Example 2 and Comparative example was operated at 50 °C and 20 mA to determine the period of half decay, based on the initial light flux. The results are shown in Table 1.

13

[0035]

Table 1

Sample	Period of half decay
Example 1	27,200
Example 2	31,100
Comparative example	5,100

[0036]

It is to be understood that a white LED exhibiting longer operating life can be provided in the present invention, as described above.

[BRIEF DESCRIPTION OF THE DRAWINGS]

[0037]

[Fig. 1] A schematic diagram showing an aerosol deposition film-forming apparatus.

[Fig. 2] A schematic cross-sectional view showing a white LED of the present invention.

[EXPLANATION OF NUMERALS]

[0038]

- 1 High-pressure gas cylinder
- 2, 6 Pipe
- 3, 5 Valve
- 4 Aerosol generation container
- 7 Chamber
- 8 Nozzle

14

- 9 Holder
- 10 Supporting substrate
- 11 XYZ θ stage
- 12 Particle raw material
- 21 White LED
- 23 Electrode
- 25 Bump
- 26 Phosphor layer
- 27 Sealing layer

FIG. 1

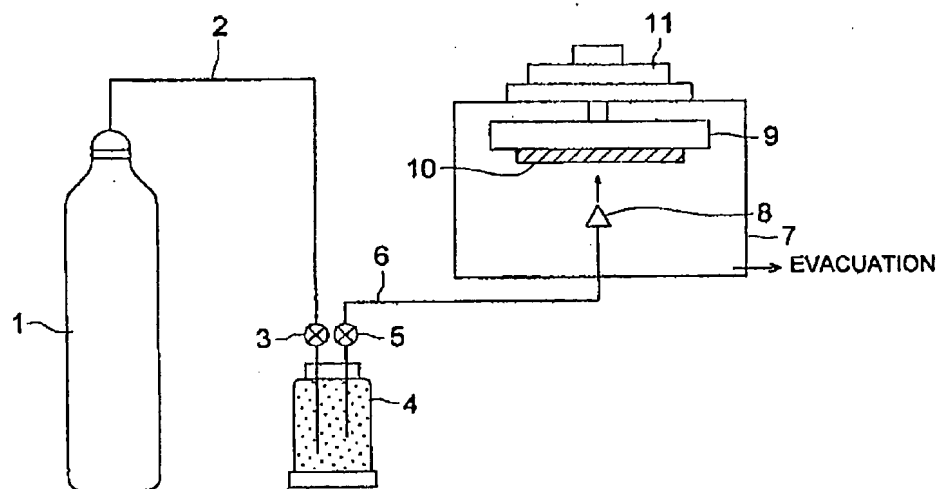
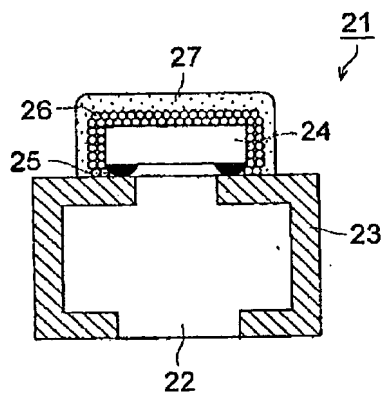


FIG. 2



1

[NAME OF THE DOCUMENT] ABSTRACT

[SUMMARY]

[OBJECT] A white LED exhibiting high reliability and longer operating life is provided by utilizing a blue LED.

[MEANS TO ATTAIN THE OBJECT] A white LED possessing a phosphor layer to convert blue light into yellow light, provided on a blue LED, wherein the phosphor layer possesses a phosphor.

[SELECTED DRAWING] None